

Development of High Stability Cathodes for Molten Carbonate Fuel Cell Applications

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Introduction

System efficiency and cost of the MCFC is advantageous when compared to other low temperature operating fuel cells. However, solubility of nickel oxide cathode in the electrolyte is one of the important problems facing the commercialization of MCFC technology. Nickel oxide reacts with the dissolved CO₂ in the electrolyte according to an acidic dissolution mechanism and dissolves in the molten carbonate during operation. The cation diffuses to the anode side of the electrolyte and is then reduced in the hydrogen atmosphere to metallic nickel. Precipitates of metallic nickel have been found in the electrolyte tile much closer to the anode and this precipitate acts as a sink for further deposition of nickel particles. This growth of the nickel layer eventually leads to a short circuit between the anode and cathode. The dissolution is accelerated under higher CO₂ partial pressure resulting in a decrease of the operating life of the cell. The dissolution of NiO also results in a decrease of the active surface area available for the oxygen reduction reaction (cathodic reaction) resulting in degradation in the cell performance.

More basic molten carbonate melts such as Li/Na carbonate eutectic does not favor the NiO dissolution process and have been used to decrease the Ni dissolution rate in the melt^{1,2}. Also, alkaline earth metal salts based on Ba or Sr have been used as additives to increase the basicity of the melt. However, using more basic molten carbonate melts only partially solves the problem, since these melts only decrease the NiO dissolution rate by 10 to 15%.

The other approach to counter the nickel dissolution problem is to either modify NiO or to identify alternate cathode materials, which have longer life in the melt. Porous electrodes with good electronic conductivity, chemical stability and proper microstructure therefore offer the excellent choice for MCFC cathodes. Several ceramic materials with small solubility in the melt have been studied as alternatives to NiO. LiFeO₂ and LiCoO₂³ offered initial promise as replacement material for NiO cathodes. LiNiO₂ possess good conductivity and has been studied for high temperature applications. However, lithium loss occurs during heat treatment and the associated stoichiometry changes are undesirable. We have adopted the solid state synthesis route to synthesize different cobalt doped LiNi_xCo_{1-x}O₂.

The performance of LiNi_xCo_{1-x}O₂ electrodes and bare nickel oxide electrodes was studied using a variety of electrochemical and physical characterization techniques.

Experimental

LiNi_xCo_{1-x}O₂ was prepared using solid-state synthesis procedure from nitrate and oxalate precursors. Porous LiNi_xCo_{1-x}O₂ was made by tape casting and sintering. Extensive electrochemical and physical characterization studies have been done to understand the performance of LiNi_xCo_{1-x}O₂ cathodes.

Results and Discussion

Sintered LiNi_xCo_{1-x}O₂ exhibits good pore structure and cathode polarization studies show low polarization indicating LiNi_xCo_{1-x}O₂ to be a promising cathode material. Figure 1 show the SEM graphs of LiNi_xCo_{1-x}O₂ with x varying from 0.7 to 0.9. Figure 2 shows the microstructure of sintered LiNi_xCo_{1-x}O₂.

Detailed characterization and performance evaluation studies will be presented during the conference.

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References

1. K. Tanimoto, Y. Miyazaki, M. Yanagida, S. Tanase, T. Kojima, N. Ohtori, H. Okuyama, T. Kodama, *Denki Kagaku* **59**(7) (1991) 619-622.
2. K. Tanimoto, Y. Miyazaki, M. Yanagida, T. Kojima, N. Ohtori, T. Kodama, *Denki Kagaku*, **63**(4) (1995) 316-318.
3. L. Giorgi, M. Carewska, S. Scaccia, E. Simonetti, F. Zarzana, *Denki Kagaku*, **64**(6) (1996) 482-485.
4. T. Fukui, S. Ohara, H. Okawa, T. Hotta, M. Naito, *J. Power Sources*, **86** (2000) 340-346.
5. J. A. Prins-Jansen, J. D. Fehribach, K. Hemmes, and J. H. W. de Wit, *J. Electrochem. Soc.*, **143**, 1617 (1996).
6. Anand Durairajan, Bala Haran, Branko Popov and Ralph White, *Journal of Power Sources*, **83**, 114 (1999).